NEW DATA ON ALPHA DECAY OF AMERICIUM ISOTOPES

S. A. BARANOV, V. M. KULAKOV, and V. M. SHATINSKIĬ

Submitted to JETP editor June 13, 1963

J. Exptl. Theoret. Phys. (U.S.S.R.) **45**, 1811-1818 (December, 1963)

Data from an investigation of the radioactive decay of Am^{243} and Am^{241} are presented. New α groups, some belonging to Am^{243}, are discovered by analyzing the α spectra. Possible identifications of newly discovered Np^{239} and Np^{237} energy levels are discussed; the existence of new $3/2^-[521]$ and $3/2^+[651]$ rotational bands is suggested, and some levels are assigned to the octopole class in the schemes of these odd-even nuclei.

INTRODUCTION

INFORMATION regarding the energy levels of Np^{239} can be obtained by studying the β decay of U^{239}, which has a short half-live of 23.5 min, and the α decay of Am^{243} with $T_{1/2} = 7950$ years. However, U^{239} is not amenable to a thorough investigation of its decay scheme. The available information regarding Np^{239} levels has been obtained mainly by analyzing the Am^{243} α spectrum, in which only five α groups with intensities $\gtrsim 0.16\%$ have heretofore been discovered. [1] Certain data concerning the α decay of Am^{243} and Am^{241} indicate that the decay schemes of these isotopes are almost identical. Our recent investigation of the Am^{241} α spectrum [2] revealed 18 groups in the α fine structure of this isotope. The α decay of Am^{243} should present a similar picture.

We have heretofore known nothing about the Np^{239} and Np^{237} levels above the excitation energies ~ 200 and 500 keV, respectively. An investigation of the low-energy α spectrum of the aforementioned americium isotopes could possibly reveal groups representing transitions to octopole levels of neptunium. Certain methodological improvements permitted the expectation that further investigation of $Am^{243,241}$ α decay would yield new information about the energy levels of the odd-even nuclei Np^{239} and Np^{237}. We employed the same apparatus that we described in [3,4].

1. PREPARATION OF RADIOACTIVE SOURCES

The Am^{243} and Am^{241} sources for the α spectroscopic investigation were prepared by vacuum evaporation of americium chlorides on thin ($\sim 0.05\,\mu$) Al_2O_3 backings. We used americium solutions containing two different isotopic concentrations of Am^{243} and Am^{241}, one of which contained 85% Am^{243} by weight, while the other contained practically no Am^{243}. These sources thus enabled unambiguous identification of α lines belonging to Am^{243} and Am^{241}.

Several sources were prepared. In the case of the preparation enriched in Am^{243} we had the maximum source area $S_{max} = 3 \times 60$ mm with the surface density $\rho = 5\ \mu g/cm^2$, and $S_{min} = 1 \times 50$ mm with $\rho = 1\ \mu g/cm^2$. The α spectrum of Am^{241} was investigated with fresh sources prepared from a solution not containing Am^{243}.

2. INVESTIGATION OF Am^{243} AND Am^{241} ALPHA DECAY

As already mentioned, the α decay of Am^{243} was investigated with a source that was not isotopically pure. The Am^{241} admixture, having a half-life 19 times shorter than that of Am^{243}, caused 75% of the total number of decays in this source. A preliminary analysis also showed that the sample contained an admixture of Am^{242m}. One decay mode of this isomeric state (with $T_{1/2} = 152$ years) is α decay comprising only 0.48% (with a partial half-life of 3.2×10^4 years) of the total decay. [5]

We estimated that the α emission from Am^{242m} could contribute not more than a few hundredths of one percent to the total activity of the source. However, the existence of the indicated admixtures would have prevented a unique identification of all the newly observed α lines of Am^{243} even if they had been clearly resolved. In addition, some of the Am^{243} α lines could coincide accidentally with Am^{241} groups, thus hindering their identification. Therefore the study of the Am^{243} α spectrum was accompanied by a continued investigation of Am^{241} α decay; in the latter case the source contained practically no admixture of Am^{242m} or Am^{243}. Special attention was devoted to analyzing the α spectra of Am^{241} and Am^{243} in the energy regions 5150—4350 and 5150—4650 keV, respectively.

Our energy standard was the most intense Am^{241} α group having the absolute energy 5468.0 ± 0.9 keV, which was recently measured by Chin Fan Leang with the magnetic α spectrograph of the Nuclear Spectroscopy Laboratory at Orsay, France (private communication).

In our experimental work the energies of the separate α groups were measured within 1 keV in the region 5500—5100 keV and not more than 2—3 keV in the region 5000—4350 keV. In the low-energy spectrum the background of scattered α particles was $(3-5) \times 10^{-7}$ particles in a 10-keV interval. This enabled us to observe low-intensity α groups having a partial half-life of $\sim 10^9-10^{10}$ years.

Figures 1 a—c show the spectrograms of Am^{243} and Am^{241} α rays obtained in three different runs with sources enriched in Am^{243}. The symbols α^{41}, α^{42}, and α^{43} indicate α groups belonging to Am^{241}, Am^{242m}, and Am^{243}, respectively. For Am^{243} the lower index is the number of the group. Figure 1a shows the strongest α lines of Am^{241} and Am^{243}. The spectrum in Fig. 1b clearly shows the most intense (91%) group α^{42}_{340} Am^{242m} at 5208 keV.[6]

Figure 1c shows a portion of the Am^{243} and Am^{241} spectrum in the region 4650—5150 keV, which contains more than 20 α groups. As already stated, to enable an unambiguous identification of the α lines we at the same time recorded the Am^{241} α spectrum in a hitherto almost uninvestigated energy range with a source containing practically no other americium isotope; this α spectrum is shown in Fig. 1d. A comparison of the last two spectra (c, d) reduced to identical intensity based on Am^{241} α emission enabled a unique assignment of the observed α groups to the correct americium isotopes. An analysis of the spectra confirmed the foregoing hypothesis that individual α groups of Am^{241} and Am^{243} are superimposed; this is clear from the spectral regions around 5100 keV in both spectrograms.

Using a source containing only Am^{241}, we also investigated the spectrum of this isotope in the region 4350—4840 keV, which is not shown in Fig. 1. In this case we were unable to resolve clearly any new α groups with intensities $\geq 2 \times 10^{-5}$% in addition to the previously observed groups α^{41}_{723} and α^{41}_{758}.

We have thus been able to observe 15 α groups in the Am^{243} spectrum. Ten of the groups were here observed for the first time; six of these are located in the previously uninvestigated low-energy

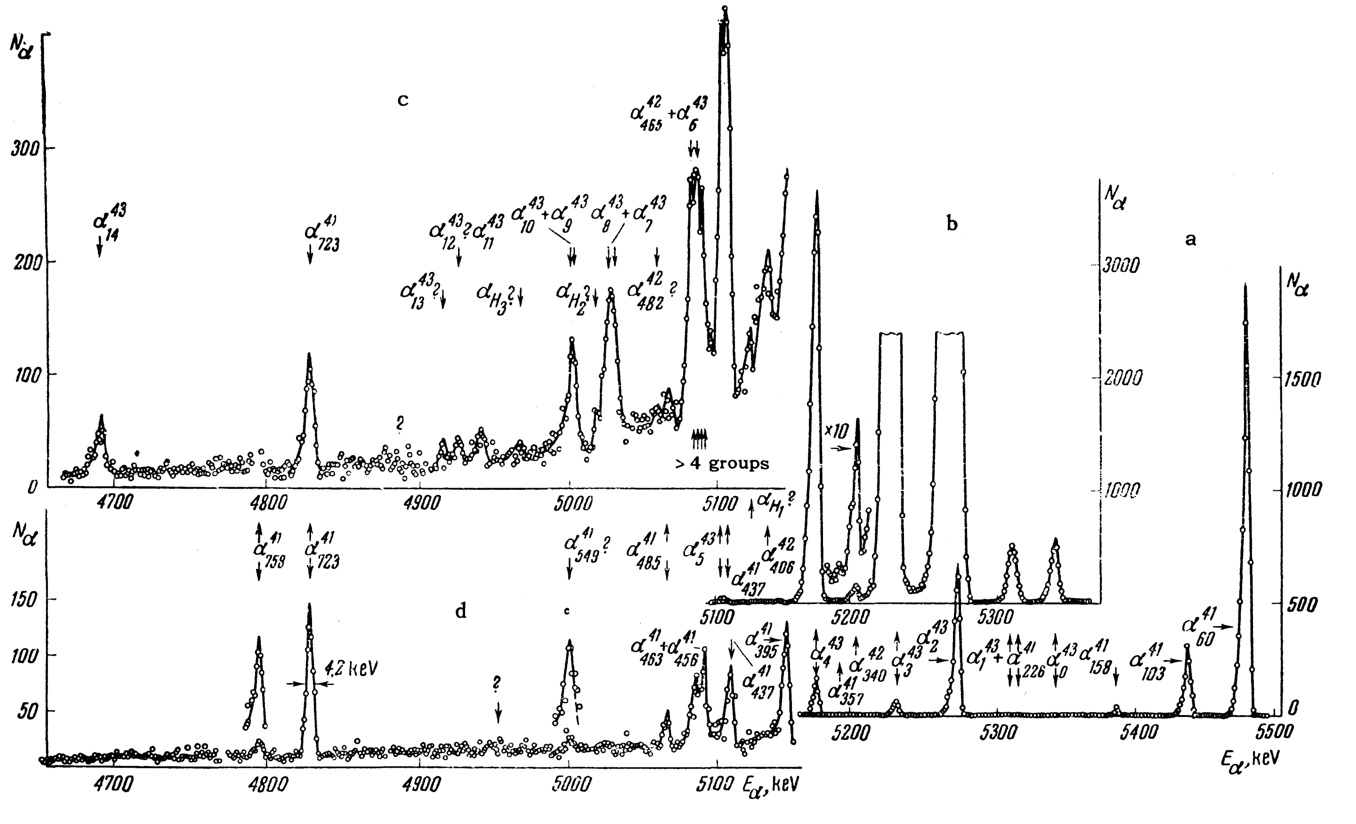

FIG. 1. Spectrograms of α radiation from $Am^{241-243}$. The ordinates represent the number of α-particle tracks per 1-mm length of the photographic plate.

Table I. Fine structure of Am^{241} α spectrum

α group	α-particle energy, keV	Intensity, %	Hindrance factor, m	Energy level, keV	α group	α-particle energy, keV	Intensity, %	Hindrance factor, m	Energy level, keV
α_0	5545	0.25	930	0	α_{10}	5194	$6\cdot10^{-4}$	3300	357
α_1	5513	0.12	1300	32.0	α_{11}	5182	$9\cdot10^{-4}$	1800	369
α_2	5486	86.0	1.3	59.6	α_{12}	5178	$3\cdot10^{-4}$	5000	373
α_3	5469	<0.04	—	76.5	α_{13}	5156	$7\cdot10^{-4}$	1700	395
α_4	5443	12.7	4.7	103	α_{14}	5114	$4\cdot10^{-4}$	1600	437
α_x	5417	$\sim10^{-2}$	—	~129	α_{15}	5096	$\sim4\cdot10^{-4}$	~1200	~456
α_5	5389	1.33	21	158	α_{16}	5089	$\sim4\cdot10^{-4}$	~1150	~463
α_6	5322	$1.5\cdot10^{-2}$	800	226	α_{17}	5068	$1.4\cdot10^{-4}$	2200	485
α_7	5279	$5\cdot10^{-4}$	13000	271	α_{18}	5004	10^{-4}	1200	549
α_8	5244	$2.4\cdot10^{-3}$	1700	306	α_{19}	4834	$7\cdot10^{-4}$	13	723
α_9	5223	$1.3\cdot10^{-3}$	2400	327	α_{20}	4800	$8.6\cdot10^{-5}$	60	758

Table II. Fine structure of Am^{243} α spectrum

α group	α-particle energy, keV	Intensity, %	Hindrance factor, m	Energy level, keV	α group	α-particle energy, keV	Intensity, %	Hindrance factor, m	Energy level, keV
α_0	5350	0.16	1900	0	α_7	5035 }	$2.2\cdot10^{-3}$	—	320
α_1	5321	0.12	1600	30.1	α_8	5029 }		—	326
α_2	5276	87.9	1.2	75.6	α_9	5008 }	$1.6\cdot10^{-3}$	1500	348
α_3	5234	10.6	5.7	118.5	α_{10}	4997? }		—	359
α_4	5181	1.1	25	172.5	α_{11}	4946	$3.4\cdot10^{-4}$	2600	411
α_5	5113	$5.4\cdot10^{-3}$	1950	241	α_{12}	4930	$1.8\cdot10^{-4}$	3700	427
α_6	5088	$4\cdot10^{-3}$	1900	267	α_{13}	4919	$8.5\cdot10^{-5}$	7000	438
					α_{14}	4695	$6.4\cdot10^{-4}$	24	666

α spectrum of Am^{241} and confirm the existence of certain Am^{242m} α lines that had been indicated in [6].

Our data are summarized in Tables I and II, which give the energies and intensities of the Am^{241} and Am^{243} α groups, the calculated hindrance factors m, and the energy levels of Np^{237} and Np^{239}.

3. ENERGY LEVEL SCHEMES OF Np^{237} AND Np^{239}

Before discussing the level schemes we shall make some comments regarding the data in Table I. Some slight disagreement exists between our present Am^{241} α-group energies and those previously reported in [2]. The explanation for this discrepancy lies in the fact that the energy standards had been the different unrelated absolute energies of the strongest α groups of Pu^{238} (E_{α_0} = 5495 keV) and Am^{241} ($E_{\alpha_{60}}$ = 5486 keV).

We believe that we made the most accurate choice in using the α_{60} line of Am^{241}, previously unknown to us, as an energy standard. It must also be mentioned that the table of the Am^{241} α fine structure published in [2] contains an unfortunate error; the calculated hindrance factors beginning with the α_{270} group are too small by a factor of 10.

In our present work we have effected a considerable reduction of the scattered-particle background, and have measured the low energy Am^{241} α spectrum (~4650—5150 keV) with energy resolution that was 1.5 times better than that in our previous work. These improvements enabled us to observe weak α^{41}_{485} and α^{41}_{549} groups in the previously investigated energy region, and to show the existence of only two levels, at 456 and 463 keV, instead of the three levels at 452, 458, and 464 keV that were given in [2]. We were able to compile a more accurate table for Am^{241} for these and other reasons.

The foregoing does not lead to any important change in the Np^{237} level scheme given in [2], so that there is no need to repeat the identification of most of these levels. However, our new data enabled us to enlarge the level scheme. Let us consider the α groups representing transitions to the following Np^{237} levels: 438, 485, 549, 723, 758, and 357 keV (Figs. 1, 2 and Table I). The first three levels can apparently be assigned to a single rotational band of Np^{237} based on a $3/2^-[521]$ orbital. The Bohr and Mottelson interval rule for odd nuclei is obeyed and a simple calculation yields $\hbar^2/2J \approx 9.4$ keV. This is consistent with the Nilsson diagram for odd Z, which predicts a $3/2^-[521]$ state.

The next two of the given levels are of great interest but cannot be described on the basis of Nilsson diagrams. It must first be mentioned that

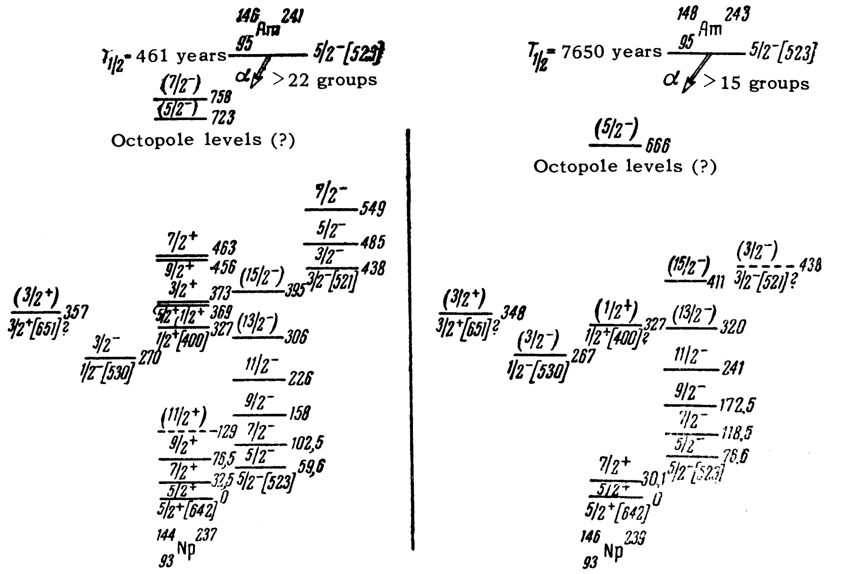

FIG. 2. Level schemes of Np^{237} and Np^{239}.

Table III

	$^3/_2{}^+$ [651]	$^1/_2{}^-$ [530]	$^5/_2{}^+$ [642]	$^5/_2{}^-$ [523]	$^1/_2{}^+$ [400]	$^3/_2{}^-$ [521]	$^5/_2$— $^5/_2$
Np^{237}	357	≲270	0	60	327	438	723
Np^{239}	348?	≲267	0	77	327?	438?	666

the hindrance factors for α transitions to the 723- and 758-keV levels are very small (having the values 13 and 60, respectively), thus evidently indicating the collective character of these levels.

The relatively large intensities, 7×10^{-6} and 8.6×10^{-7}, of the α transitions to these two levels favor the assignment of octopole symmetry similar to the 1- and 3- levels in neighboring even-even nuclei. An analysis of the very scanty experimental data regarding the position of the first level of a rotational band having octopole symmetry in heavy nuclei also favors the attribution of octopole symmetry to these levels. The quantum characteristics (J, π, K) of these levels should evidently be ($^5/_2$—$^5/_2$) and ($^7/_2$—$^5/_2$). If this is correct it can easily be shown that the constant in Bohr and Mottelson's formula for odd-mass energy levels is $\hbar^2/2J = 4.9$ keV. Therefore the moments of inertia of the Np^{237} nucleus in the ground state and the considered state agree within experimental error.[1)]

The Np^{237} level scheme in Fig. 2 also contains a 357-keV level. This could be the base of a $^3/_2{}^+$[651] band which should be present in this level scheme according to the Nilsson diagram.

The 22 α groups observed in the Am^{241} α spectrum now enable us to conclude that at least 7 bands exist in the Np^{237} level scheme, one of which results from octopole oscillations of this nucleus.

Table III shows the orbitals and energies on which these bands are built in Np^{237} and Np^{239}; we shall now discuss the latter nucleus. The level scheme of Np^{239} is shown in Fig. 2. The experimental data indicate that this nucleus possesses more than 15 levels in the energy region 0—700 keV. The ground state of Np^{239}, like those of all other neptunium isotopes, has spin $^5/_2$ [7] and positive parity. Very great similarity is easily observed when the strongest α groups of Am^{241} and Am^{243} and their energy separations are compared. This appears to indicate that the decay schemes of these isotopes are almost identical. The Np^{239} ground state and 77-keV excited state have the respective assignments $^5/_2{}^+$[642] and $^5/_2{}^-$[523].[8,9] Figure 2 shows that the favored $^5/_2{}^-$[523] band is highly developed; we were able to observe 6 of its members with the successive spins and parities, $^5/_2{}^-$, $^7/_2{}^-$, $^9/_2{}^-$, $^{11}/_2{}^-$, $^{13}/_2{}^-$, and $^{15}/_2{}^-$.

If we could reason by analogy, the excited 327-, 348-, and 438-keV levels of Np^{239} would be given the assignments $^1/_2{}^+$[400], $^3/_2{}^+$[651], and $^3/_2{}^-$[521]. The 267-keV level appears to be the second member of the $^1/_2{}^-$[530] band with spin $^3/_2{}^-$. The addition of two neutrons to the Np^{237} nucleus must, of course, affect the level structure of the odd-mass nucleus Np^{239} and reasoning by analogy is not an ideal procedure. We therefore do not insist that

1) $\hbar^2/2J = 4.75$ keV for the ground state of Np^{237}.

our identifications of the newly observed levels must be correct; we regard our foregoing assignments as providing only a working hypothesis.

The 666-keV level is very interesting. The small hindrance factor 24 for an α transition to this level (the α_{666}-group intensity is $\sim 8 \times 10^{-6}$) also indicates that it belongs to the class of octopole levels. It lies, to be sure, about 60 keV below the corresponding level of Np^{237}, but this is not at all surprising, since we know that the energies of this class of levels depend on the values of A and Z for a specific nucleus. The 666-keV level must apparently have the quantum assignment $5/2-5/2$.

It is of some interest to note that in [10], where γ radiation from a (n, γ) reaction in U^{238} was investigated, it is indicated that U^{239} possesses octopole levels at 665 and 690 keV. The first of these energies agrees almost exactly, within experimental error, with the octopole levels of Np^{239} observed by us. It is difficult to decide on the basis of these two experiments alone whether this close energy agreement of the two octopole levels is only accidental.

The number of levels in the Np^{239} scheme has not been exhausted by the foregoing. The spectrum shown in Fig. 1c appears to indicate additional α groups, three of which have been designated by α_H. We have not identified these α groups and the corresponding levels are not shown in Fig. 2.

In conclusion we wish to thank N. I. Aleshin, A. A. Arutyunov, Yu. N. Dmitriev, and K. I. Merkulova, who assisted us with the measurements. A. P. Smirnov-Averin for furnishing the Am^{243} sample, and L. V. Chistyakov and G. I. Khlebnikov for the careful supplementary removal of impurities from the americium samples.

[1] Stephens, Hummel, Asaro, and Perlman, Phys. Rev. **98**, 261A (1955).

[2] Baranov, Kulakov, Shatinskiĭ, and Zelenkov, JETP **43**, 795 (1962), Soviet Phys. JETP **16**, 562 (1963).

[3] Baranov, Zelenkov, Shchepkin, Beruchko, and Malov, Atomnaya énergiya **7**, 262 (1959), Soviet J. Atomic Energy **7**, 756 (1959); Izv. AN SSSR **23**, 1402 (1959), Columbia Tech. Transl. p. 1389.

[4] Baranov, Zelenkov, and Kulakov, Izv. AN SSSR **24**, 1035 (1960), Columbia Tech. Transl. p. 1045.

[5] Barnes, Henderson, Harkness, and Diamond, J. Inorg. Nuclear Chem. **9**, 105 (1959).

[6] Asaro, Hichel, Thompson, and Perlman, UCRL-9566, 32, 1961.

[7] J. C. Hubbs and R. Marrus, Phys. Rev. **110**, 287 (1958).

[8] B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Skrifter **1**, No. 8 (1959).

[9] Bohr, Fröman, and Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **29**, No. 10 (1955).

[10] Emergy, Fiebiger, and Kane, Bull. Am. Phys. Soc. **7**, 11 (1962).

Translated by I. Emin
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ERRATUM (published in JETP **46**, February 1964)

It is suggested in this article that the 723- and 758-keV levels in the Np^{237} scheme and the 666-keV level in Np^{239} correspond to octopole oscillations of the Np nucleus. However, a recent (27 December 1963) letter from O. Nathan reports that Asaro, Lederer, and Perlman also observed a 723-keV level in Np^{237} by a coincidence method. They estimate an E0 multipolarity for the γ transition ($E_\gamma \approx 660$ keV) between the 723-keV level and the $5/2^-$ [523] state ($E_{lev} \approx 60$ keV). This gives grounds for assuming that the 723-keV level corresponds to β oscillations of Np^{237} from the $5/2^-$ [523] state. The 666-keV level which we found in Np^{239} is apparently of the same nature. It should be noted that such an interpretation of these levels does not contradict our experimental data.